Ultrafast structural dynamics of highly excited nanoparticles in intense laser fields

Recent developments of high-intensity femtosecond lasers generated by chirped pulse amplification have given us new opportunities to transform matter to novel states under extreme conditions. Intense and ultrashort infrared laser pulses deposit energy into matter with a femtosecond exposure time, thus triggering nonequilibrium dynamic processes such as the creation of a dense and transient electron-hole plasma. During the creation of the plasma, marked structural changes in the matter are expected to occur. Elucidating how and on what timescale structural changes proceed in the matter under the intense laser field leads to a deep understanding of the fundamental mechanisms of excitation and deexcitation in highly excited systems.

For the study of the fundamental aspects of laser-matter interaction, atomic clusters, especially noble-gas clusters, have been used as a testbed [1] owing to their characteristics such as ease of generation, size tunability, and an isolated system of bulk density. Numerous studies have revealed that a nanoscale atomic cluster exposed to an intense near-infrared (NIR) laser pulse becomes a nanoscale plasma (nanoplasma) [2]. Thus far, the properties and dynamics of laser-induced nanoplasmas have been studied by ion, electron, and fluorescence spectroscopies, by which one can only infer the structural changes in clusters.

The emergence of XFELs has changed the situation; the XFEL enables us to investigate the structural dynamics of nanoscale systems with an atomic-scale spatial resolution and a temporal resolution of several-ten-femtoseconds [3], using the diffraction-before-destruction scheme [4].

In this work, we investigated ultrafast structural dynamics in xenon (Xe) clusters irradiated by the NIR laser, employing the time-resolved wide-angle X-ray scattering (WAXS) technique. The experiments were carried out at experimental hutch 2 of SACLA **BL3** [5]. A schematic of the experiment is depicted in Fig. 1. A jet of Xe clusters (~1×10^7 atoms/cluster) was irradiated by a NIR laser pulse (1.55 eV, 30 fs FWHM, 4×10^{16} W/cm^2) followed by a single XFEL pulse (11 keV, 10 fs FWHM, 4×10^{17} W/cm^2) at a selected temporal delay. The temporal delay of the XFEL pulse relative to the NIR pulse was controlled by using the delay stages installed to the NIR laser system. The temporal jitter between the arrival time of the two laser pulses was measured using the arrival timing monitor [6]. Scattering signals from Xe clusters were collected by a short-work-distance multiport CCD detector for each XFEL shot. Xe ion signals were collected by an ion time-of-flight (TOF) spectrometer.

The experimental results are summarized in Fig. 2. We chose intense Bragg spots observed at each delay dataset for analysis to ensure the good exposure condition between XFEL pulses and clusters. Clear changes in the profiles of Bragg spots from the fcc (111) reflection of single Xe nanoparticles could be observed by applying this data filtering. As shown in Fig. 2(a), bright Bragg spots were observed before NIR irradiation and their intensity decreased after NIR irradiation. Figures 2(b) and 2(c) show the qualitative temporal development of the intensity and width (FWHM) of the Bragg spots with respect to the XFEL delay. The spot intensity decreased after NIR irradiation, while the spot width increased by a few tens of percent. The simultaneous behavior could be

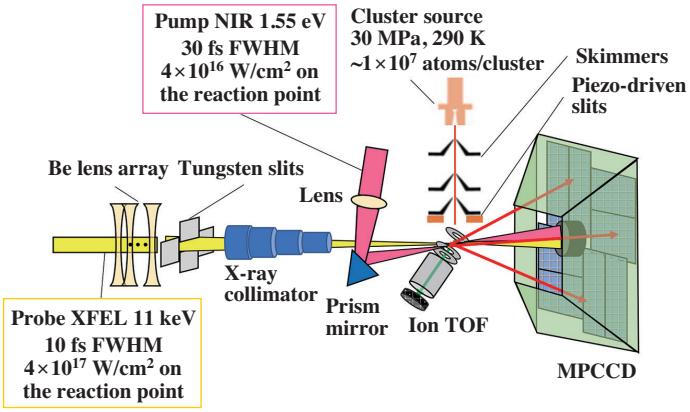

Fig. 1. Schematic of the time-resolved experiments at SACLA BL3. [5]

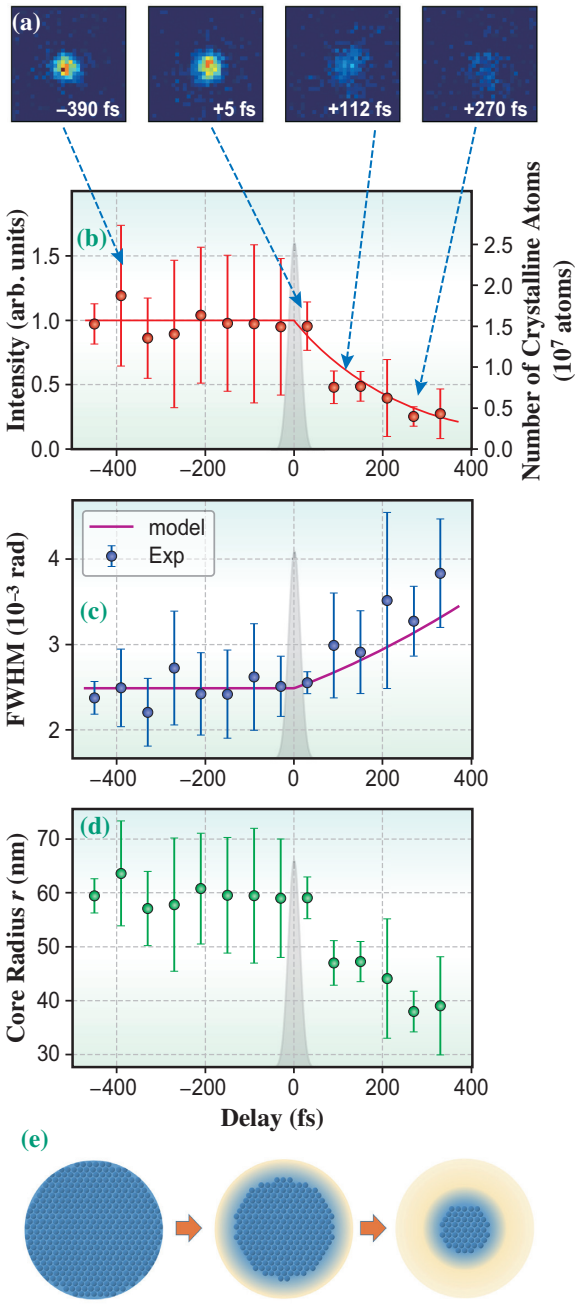

Fig. 2. Delay dependence of profiles of Bragg spots from fcc (111) reflection of xenon nanoparticles. **(a)** Characteristic spot images. **(b)** Temporal development of the spot intensity (marker). **(c)** Temporal development of the spot width (marker). **(d)** Temporal development of the radius of the crystalline core (marker). **(e)** Schematic of the crystal disordering consistent with the current experiments. [5]

reproduced by the model shown in Fig. 2(e), where the disordering of the crystalline structure proceeds from the surface (outer part) to the core (inner part) of the Xe nanocrystal.

Furthermore, the propagation speed of disordering, which was extracted from the temporal development of the radius of the crystalline core shown in Fig. 2(d), is close to the plasma sound speed estimated from the ion kinetic energy distributions. The present results can be interpreted as that the crystalline order of the core initially survives and is detectable even under an intense laser field, while local disordering on the nanoplasma proceeds from the surface towards the core with speed of the same order as the plasma sound speed.

In summary, ultrafast and atomic-scale structural changes in nanoplasma created under an intense NIR laser field were clarified by shot-by-shot time-resolved WAXS experiments at SACLA. Our findings provide a new insight into the structural dynamics in nanoplasma formation and its development on the atomic-scale spatial resolution and temporal resolution of several tens of femtoseconds.

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